Air Mass Characterisation During EOPACE: Aerosol Composition and Concentration

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Award No. N00014-96-1-0321
http://sunspot.nosc.mil:80/543/eopace/eomain.html

LONG-TERM GOAL

This project aims to define those aerosol properties within the littoral zone which impinge upon the performance of Navy electro-optical systems for a wide range of environmental conditions. These surface observations are being utilised to validate aerosol optical depths and limited particle size information derived from satellite imagery at multiple wavelengths. It is anticipated that these observations of aerosol composition and concentration aerosol observations will assist with the development and validation of mesoscale models incorporating aerosol sources, sinks and transport.

OBJECTIVES

The major objective of the current work has been to characterise the composition and concentration of the accumulation mode particles ($0.05 < r < 1.5\mu m$), for a variety of locations and environmental conditions within the littoral zone, by means of a thermal analytical technique. Soot carbon loadings have been measured within these coastal air masses both as a cross-check on the thermal analytical analysis and as a very useful means of defining the anthropogenic input into these air masses. In addition, the impact of size-differentiated aerosol composition upon propagation at visible and infra-red wavelengths is being examined.

APPROACH

The thermal analytical ('volatility') approach employed to determine the concentrations and composition of sub-micron aerosol particles consists of a Particle Measuring Systems ASASP-X optical particle counter, covering particle radii from 0.05 to 1.5μm, to which particles are supplied from a quartz heater tube capable of cycling through a temperature range from near ambient to close to 1000°C. The method relies upon the fact that most aerosol species become volatile at a characteristic temperature enabling the predominant components of the aerosol to be identified. The volatility technique has been used previously in the littoral zone (Smith and O'Dowd, 1995) where the ability to derive soot carbon measurements from the thermal response of the aerosol was demonstrated. However, because of the importance of these soot carbon measurements in indicating the impact of

Report Documentation Page

Form Approved
OMB No. 0704-0188

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1. REPORT DATE **1998**	2. REPORT TYPE	3. DATES COVERED **00-00-1998 to 00-00-1998**
4. TITLE AND SUBTITLE **Air Mass Characterisation During EOPACE: Aerosol Composition and Concentration**		5a. CONTRACT NUMBER 5b. GRANT NUMBER 5c. PROGRAM ELEMENT NUMBER
6. AUTHOR(S)		5d. PROJECT NUMBER 5e. TASK NUMBER 5f. WORK UNIT NUMBER
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) **University of Sunderland,Centre for Marine and Atmospheric Sciences,Benedict Building, St Geoerge's Way,Sunderland SR2 7BW, UK,**		8. PERFORMING ORGANIZATION REPORT NUMBER
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)		10. SPONSOR/MONITOR'S ACRONYM(S) 11. SPONSOR/MONITOR'S REPORT NUMBER(S)
12. DISTRIBUTION/AVAILABILITY STATEMENT **Approved for public release; distribution unlimited**		
13. SUPPLEMENTARY NOTES **See also ADM002252.**		
14. ABSTRACT		
15. SUBJECT TERMS		

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT **unclassified**	b. ABSTRACT **unclassified**	c. THIS PAGE **unclassified**	**Same as Report (SAR)**	**6**	

anthropogenic (principally vehicular and power plant) inputs within coastal air masses, a Magee Scientific aethalometer was deployed to measure this parameter independently.

Particles larger than about 1μm in radius were measured using two additional Particle Measuring Systems aerosol counters: an FSSP-100 and an OAP-230X covering particle radius ranges from 0.25 to 23.5μm, and 5 to 150μm, respectively. For the shore-based campaigns, these observations are being used in associated studies (and in combination with additional measurements) to define the surf zone contribution to littoral aerosol loadings but, in all the campaigns, they provide information on the complete particle size range of interest for infra-red propagation studies.

WORK COMPLETED

The very substantial quantities of data obtained from 7 EOPACE field projects (Jensen, 1995) during 1995-97 have now been completely analysed and supplied to the EOPACE community. Specific periods from these data coinciding with satellite overpasses have been validated in more detail and supplied to Dr Wash and Ms Jordan at the Naval Postgraduate School for comparison with satellite-derived optical depths and estimates of the particulate size distribution (Jensen *et al*, 1998). The sensitivity of atmospheric propagation at visible and infra-red wavelengths of interest to aerosol composition has been examined for a subset of these data obtained on Scripps Institution of Oceanography pier during 1997.

RESULTS

The analysis of aerosol data supplied to associated participants in the EOPACE Programme has been reported elsewhere (De Leeuw *et al*, 1997, Gathman & Smith, 1997, Smith & Hill, 1997, Veefkind *et al*, 1997) and this report concentrates upon the impact of differing aerosol composition upon atmospheric extinction at visible and infra-red wavelengths, using observations made on Scripps pier during the period from 31March to 12 April 1997, for which the meteorological conditions were characterised by strong land-sea breeze influences. Typically, overnight winds would give a light, easterly flow with velocities of about 2m s^{-1}, sometimes less than 1m s^{-1}, which would switch quite rapidly to the north west in mid-morning. These westerly winds would freshen to perhaps 5m s^{-1}, or more, before slackening and turning easterly in the evening. During these easterly periods, the local air flow is driven by nocturnal cooling over the land with a stable, subsiding air mass passing offshore and giving sharply-reduced air temperatures. Offshore flow brought polluted air from the urban areas around the site, giving soot carbon loadings in excess of 1000ng m^{-3} at such times. Although westerly, or northwesterly, wind directions generally brought cleaner air to the measurement site, considerable hysteresis was noted in the system. The low settling velocities of these smaller aerosol particles means that loss rates are generally quite low, with atmospheric residence times for conditions typical of this project being several days or more. Thus, polluted air flowing offshore during the periods of easterly winds would lose little of its aerosol content before returning across the littoral zone on subsequent westerly winds. On occasions, stronger westerly air flows would bring in relatively clean oceanic air with soot carbon loadings below about 40ng m^{-3}. These changes in aerosol composition and concentration were clearly evident in the volatility analysis of aerosol composition.

To these smaller aerosol particles, transported relatively unchanged within a given air mass, were added large particles produced by whitecapping at the ocean surface (due to the wind over the open ocean, or by breaking waves in the surf zone) or as wind-blown dust from the land. Often, during periods of nocturnal offshore flow, an internal boundary layer formed over the surf zone driven by the injection of water vapour from the ocean surface (and from the sea spray droplets produced in the surf) which maintained its buoyancy relative to the cooler, stable air aloft due to the lower density of moist air. The dramatic injection of moist plumes into this internal boundary layer, which contrasted sharply with the crystal clear air only a few metres above, was clearly evident on these occasions.

Measurements from the various particle counters were combined into hourly averages to provide spectra covering the radius range from 0.05 to 150µm. For the larger particles, this task consisted of simply integrating the successive one-minute spectra within each hour. However, for the smaller aerosol sizes measured by the volatility system, the procedure was complicated by the fact that, even when the heater system was at its lowest temperatures, it remained significantly warmer than the ambient environment and, consequently, these particles were substantially drier and therefore smaller than when in equilibrium with the atmosphere. In order to overcome this difficulty, the 5-second spectra recorded by the volatility system for temperatures near ambient (< 60°C) were integrated for each hour and then 'grown' to their equilibrium sizes at the prevailing atmospheric relative humidity using a simple parameterisation (Gerber, 1988).

Fifth-order polynomial were fitted to these hourly-averaged spectra to smooth the statistical noise between the aerosol probe channels, with the atmospheric extinction at four wavelengths being calculated from this polynomial by means of a Mie scattering algorithm (Bohren and Huffman, 1983). To simplify the atmospheric extinction calculations, single representative values of the complex refractive index at each wavelength were used for the complete aerosol spectrum, as specified for the four wavelengths in Table 1 below (following Shettle & Fenn, 1979).

Wavelength (µm)	0.55	1.06	3.5	10.6
Refractive Index Real component	1.332	1.367	1.423	1.271
Refractive Index Imaginary component	-5.83E-09	-9.85E-09	-5.64E-03	-4.17E-02

Table 1: Wavelengths and refractive indices used for atmospheric extinction calculations

The variations in atmospheric extinction at these four wavelengths were computed for the whole measurement period and demonstrated the variability of extinction for the diverse changes in aerosol loadings over the period, as shown in Figure 1. While the major features of these time series show a strong correlation across all wavelengths, the ratios of the extinctions at different wavelengths are far from constant, for example the ratio of the extinction at 0.55µm to that at 10.6µm varies from less than 2 to more than 12 over the period in question. A number of case studies were selected for more detailed examination on the basis of the criteria including soot carbon loading, visibility and the ratio of extinction at 10.6µm to that at 0.55µm.

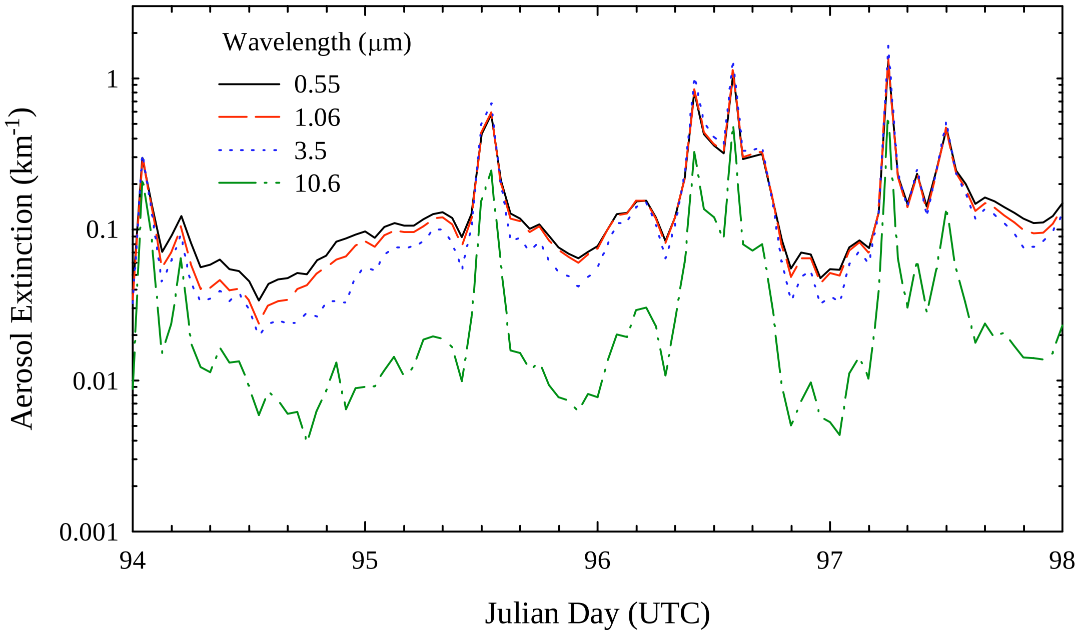

Figure 3: Atmospheric extinction over the period from 4 to 8 April 1997 (JD 94 - 98)

To determine whether atmospheric extinctions at the various wavelengths were affected significantly by the assumptions made regarding the complex refractive indices of the aerosol particulates, revised indices were derived based upon the specific compositions of the particles for each of the case studies. The particle spectra were split into two components - the small particles for which relatively detailed information was available from the volatility analysis, and the larger particles which were assumed to be composed of dust particles for offshore flow, and sea salt particles for onshore flow

For the smaller aerosol particles, it was assumed that the number concentrations of the individual constituents, as determined from the composiition information, reflected the relative proportions of each material. Thus, amended refractive indices were calculated based upon a geometric weighting of the refractive indices for each material. All the aerosol particles were assumed to be hygroscopic and, under the ambient relative humidity conditions typical of this project, the particles would be approximately twice the diameter of their dry residues (i.e. about 85% of the particulate volume would be water), and the refractive indices were adjusted accordingly. While much more complex weighting schemes for the aerosol mixtures could be envisaged, allowing for volume rather than number mixing for the small aerosol and internal as well as external mixing, it was felt that the approach adopted was straightforward and adequate to test the sensitivity of the atmospheric aerosol extinctions to realistic variations in particulate composition.

Generally, the refractive indices (both real and imaginary components) for the smaller aerosol particles are increased relative to the initial estimates shown in Table 1 when more detailed account is taken of their composition, leading to an increased contribution to the extinction from these particles.

Conversely, the real part of the refractive indices for the larger particulates are reduced from their Table 1 values, especially for the ‘dust-like’ offshore flow conditions, although the imaginary component is increased, especially at 10.6µm wavelength. Thus, in cases where the smaller aerosol particles are at their highest concentrations, the net result is an increased extinction at 0.55, 1.06 and 3.50µm wavelengths, whereas, in other cases, the greater extinction from the small particles is more than offset by the decreased contribution from the larger particles. However, in general, composition of the particles plays a minor role in determining their contribution to atmospheric extinction compared with that played by the size distribution, for environmental conditions of interest in electro-optical propagation. Close to the surface in littoral regions, the dominant influence upon the extinction at all wavelengths is likely to be due to sea spray particles produced both in the surf zone and by the action of the wind upon the surface, although anthropogenic inputs may contribute substantially to the extinction in relatively polluted conditions.

One matter which must be stressed is the sensitivity of the extinction calculations to the presence of low concentrations of relatively large aerosol particles. Attention is drawn to the long time scales required to obtain representative aerosol samples with current optical particle counters for all but the highest atmospheric extinctions, since most optical particle counters have volume sampling rates of no more than 20ml s^{-1}. This problem is of greatest significance when dealing with infra-red wavelengths where the contribution of the larger aerosol particles is most marked.

IMPACT/APPLICATION

These measurements are useful in defining the high degree of variability in aerosol concentrations and composition encountered within the littoral one and emphasises that point measurements are likely to be of only limited benefit in defining the air mass characteristics. However, the generally low loss rates of the accumulation mode particles which dominate air mass characteristics means that they remain mostly conserved within a given air mass (with allowances for changes in relative humidity). Hence, current mesoscale meteorological models should be capable of development to incorporate aerosol sources, sinks and transport processes in order to provide predictions of these air mass characteristics. These measurements provide a topographic map of aerosol for use in such mesoscale models.

TRANSITION

This investigation forms an element of the EOPACE programme whose aim is to characterise the atmospheric environment for the evaluation of electro-optical system performance.

RELATED PROJECTS

The UK Ministry of Defence provide funding via DERA Portsdown Contract No: SSDH300037 to cover participation in the surf zone studies which form an additional element of the EOPACE programme.

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